

Emergence of magnetic long-range order in kagome quantum antiferromagnets

Johannes Richter and Oliver Götze

Institut für Theoretische Physik, Universität Magdeburg, 39016 Magdeburg, Germany

E-mail: johannes.richter@physik.uni-magdeburg.de

Abstract. The existence of a spin-liquid ground state of the $s = 1/2$ Heisenberg kagome antiferromagnet (KAFM) is well established. Meanwhile, also for the $s = 1$ Heisenberg KAFM evidence for the absence of magnetic long-range order (LRO) was found. Magnetic LRO in Heisenberg KAFMs can emerge by increasing the spin quantum number s to $s > 1$ and for $s = 1$ by an easy-plane anisotropy. In the present paper we discuss the route to magnetic order in $s = 1/2$ KAFMs by including an isotropic interlayer coupling (ILC) J_{\perp} as well as an easy-plane anisotropy in the kagome layers by using the coupled-cluster method to high orders of approximation. We consider ferro- as well as antiferromagnetic J_{\perp} . To discuss the general question for the crossover from a purely two-dimensional (2D) to a quasi-2D and finally to a three-dimensional system we consider the simplest model of stacked (unshifted) kagome layers. Although the ILC of real kagome compounds is often more sophisticated, such a geometry of the ILC can be relevant for barlowite. We find that the spin-liquid ground state present for the strictly 2D $s = 1/2$ XXZ KAFM survives a finite ILC, where the spin-liquid region shrinks monotonously with increasing anisotropy. If the ILC becomes large enough (about 15% of intralayer coupling for the isotropic Heisenberg case and about 4% for the XY limit) magnetic LRO can be established, where the $q = 0$ symmetry is favorable if J_{\perp} is of moderate strength. If the strength of the ILC further increases, $\sqrt{3} \times \sqrt{3}$ LRO can become favorable against $q = 0$ LRO.

1. Introduction

One of the most fascinating problems in frustrated quantum magnetism is the investigation of the ground state (GS) of the $s = 1/2$ antiferromagnet on the kagome lattice, see, e.g., Refs. [1–20]. The existence of a spin-liquid ground state of the $s = 1/2$ Heisenberg kagome antiferromagnet (KAFM) is well established, although the discussion of the nature of the spin-liquid state is still not settled. Meanwhile, also for the $s = 1$ Heisenberg KAFM evidence for the absence of magnetic long-range order (LRO) was found [12, 21–24]. On the other hand, recently it has been reported that magnetic $\sqrt{3} \times \sqrt{3}$ LRO can be established by increasing the spin quantum number to $s > 1$ [12, 20, 25]. Further increasing s stabilizes this kind of order, thus a coupled-cluster study reports for the magnetic order parameter (sublattice magnetization) $M/s = 0.074, 0.203, 0.294$, and 0.358 for $s = 3/2, 2, 5/2$ and 3 , respectively [12]. Note that in [20] for $s = 3/2$ a similar value for M was reported, whereas in [25] a significantly larger M was obtained.

Another way to decrease quantum fluctuations is to include exchange anisotropy [18, 25–34] or to take into account further-neighbor couplings [31, 35–48]. Such modifications of the pure nearest-neighbor (NN) Heisenberg KAFM may play a crucial role to establish GS magnetic LRO of $\sqrt{3} \times \sqrt{3}$ or of $q = 0$ symmetry. For the XXZ $s = 1/2$ KAFM with NN couplings only, an

easy-axis anisotropy is not sufficient to establish magnetic LRO [18, 29–31, 34]. On the other hand, for $s = 1$ there is evidence [20, 28, 30] that a finite region of disorder around the isotropic Heisenberg point gives way for $\sqrt{3} \times \sqrt{3}$ LRO as increasing the easy-axis anisotropy. Further increasing the anisotropy towards the XY model yields a second transition from $\sqrt{3} \times \sqrt{3}$ to $q = 0$ LRO.

An obvious route towards magnetic LRO in the GS and also at finite temperatures is given by including a coupling between kagome layers. Here we investigate the effect of an isotropic interlayer coupling (ILC) on the ground state order in a stacked $s = 1/2$ XXZ KAFM with easy-plane anisotropy in the layers by using the coupled-cluster method (CCM) to high orders of approximation. The corresponding Hamiltonian reads

$$H = \sum_n \left\{ \sum_{\langle ij \rangle} \left(s_{i,n}^x s_{j,n}^x + s_{i,n}^y s_{j,n}^y + \Delta s_{i,n}^z s_{j,n}^z \right) \right\} + J_\perp \sum_{i,n} \mathbf{s}_{i,n} \cdot \mathbf{s}_{i,n+1}, \quad (1)$$

where n labels the kagome layers and J_\perp is a perpendicular (i.e., non-frustrated) ILC. The expression in curly brackets represents the XXZ KAFM of the layer n with NN intralayer couplings $J = 1$ and an easy-axis anisotropy $0 \leq \Delta \leq 1$. We do not restrict the sign of the ILC, i.e., we consider ferro- as well as antiferromagnetic J_\perp .

It is in order to notice that the ILC in real kagome compounds is often more sophisticated than that one considered here. Hence, the aim of our paper is not the discussion of particular kagome compounds, rather it is a theoretical investigation of the general question for the crossover from a purely two-dimensional (2D) to a quasi-2D and then to a three-dimensional system. For that it is appropriate to consider the simplest model of stacked unshifted kagome layers with a perpendicular ILC. There is at least one kagome compound, namely barlowite, with unshifted kagome layers, see, e.g. [49, 50]. As it has been pointed out very recently [51], through isoelectronic substitution in barlowite this kagome system becomes quite similar to our model system.

As already mentioned above, for isolated $s = 1/2$ kagome layers the GS is always magnetically disordered. Very recently, for the fully isotropic model it has been found that this non-magnetic ground state persists until relatively large strengths of the ILC [52], i.e., the critical strength of the ILC J_\perp^c needed to establish magnetic LRO is about 15% of the intralayer coupling. The question arises how an additional XXZ anisotropy in the layers influences J_\perp^c . Does the magnetically disordered GS survive also for the extreme limit of XY layers, if the ILC becomes nonzero? Bearing in mind that the GS selection for the strictly 2D model depends on the anisotropy parameter Δ [20, 28, 30] we may also ask which GS symmetry ($\sqrt{3} \times \sqrt{3}$ or of $q = 0$) is selected when including the ILC. Moreover, we will discuss to what extent the sign of J_\perp is relevant.

2. Brief outline of the coupled-cluster method (CCM)

The CCM is meanwhile a well-established and well-tested method in frustrated quantum magnetism, see, e.g. [12, 30, 37, 39, 52–63]. Hence, we want to illustrate here only some basic features of the CCM. At that we follow [12, 30], where the CCM was applied to the 2D KAFM, and [52], where the isotropic stacked $s = 1/2$ KAFM was studied by means of the CCM. For more general information on the CCM, see, [64–69]. Note first that the CCM yields results directly for number of sites $N \rightarrow \infty$. The starting point of the CCM calculation is a normalized reference state $|\Phi\rangle$. From a quasi-classical perspective that is the stacked coplanar $\sqrt{3} \times \sqrt{3}$ state or $q = 0$ state (see, e.g., Refs. [28, 30, 70–73]). Then we perform a rotation of the local axes of each of the spins such that all spins in the reference state align along the negative z axis. Within this rotated local spin coordinate system we define a complete set of multispin creation operators $C_I^+ \equiv (C_I^-)^\dagger$ related to this reference state:

$|\Phi\rangle = |\downarrow\downarrow\downarrow\cdots\rangle$; $C_I^+ = s_n^+, s_n^+s_m^+, s_n^+s_m^+s_k^+, \dots$, i.e., the spin operators entering the multispin creation operators are defined in the local coordinate frames. The indices n, m, k, \dots denote arbitrary lattice sites. The ket and bra GS eigenvectors $|\Psi\rangle$ and $\langle\tilde{\Psi}|$ of the spin system are given by $|\Psi\rangle = e^S|\Phi\rangle$, $S = \sum_{I \neq 0} a_I C_I^+$; $\langle\tilde{\Psi}| = \langle\Phi|\tilde{S}e^{-S}$, $\tilde{S} = 1 + \sum_{I \neq 0} \tilde{a}_I C_I^-$. For the coefficients a_I and \tilde{a}_I in the CCM correlation operators, S and \tilde{S} , the ket-state and bra-state equations $\langle\Phi|C_I^- e^{-S} H e^S |\Phi\rangle = 0$, $\langle\Phi|\tilde{S} e^{-S} [H, C_I^+] e^S |\Phi\rangle = 0$, $\forall I \neq 0$, hold, where each equation belongs to a certain multispin configuration I , i.e., to a certain configuration of lattice sites n, m, k, \dots . For the GS energy $E_0 = \langle\Phi|e^{-S} H e^S |\Phi\rangle$ holds. The sublattice magnetization (magnetic order parameter) is given by $M = -\frac{1}{N} \sum_{i=1}^N \langle\tilde{\Psi}|s_i^z|\Psi\rangle$, where s_i^z is expressed in the rotated local coordinate system. For the many-body problem at hand, we have to truncate the expansions of S and \tilde{S} . For that we use the standard LSUB m approximation scheme, cf., e.g., [12, 30, 37, 39, 52–58, 60–63, 67, 68], where no more than m spin flips spanning a range of no more than m contiguous lattice sites are included. Using the parallelized CCM code [74] we can solve the set of CCM equations up to LSUB8. For this approximation level the number of CCM equations is 278,024. We then extrapolate the ‘raw’ LSUB m data to the limit $m \rightarrow \infty$. In correspondence to [52] we use two schemes, namely an extrapolation using $m = 4, 5, \dots, 8$ (scheme I) and separately an extrapolation using $m = 4, 6, 8$ (scheme II). Scheme I corresponds to that one used for the 2D KAFM [12, 30], whereas scheme II (not including the odd LSUB m approximation levels) is more appropriate for magnets with collinear AFM correlations [53–55, 57–59, 62, 65, 75]. By comparing the results of schemes I and II we can get information on the precision of the extrapolated data.

For the GS energy the formula $e_0(m) = E_0(m)/N = e_0(m \rightarrow \infty) + a_1/m^2 + a_2/m^4$ is well tested and it provides precise data for the extrapolated energy $e_0(m \rightarrow \infty)$; for the magnetic order parameter M the ansatz $M(m) = M(m \rightarrow \infty) + b_1(1/m)^{1/2} + b_2(1/m)^{3/2}$ is an appropriate choice to determine quantum critical points for frustrated quantum spin systems [12, 30, 37, 39, 52–55, 57, 58, 62, 63].

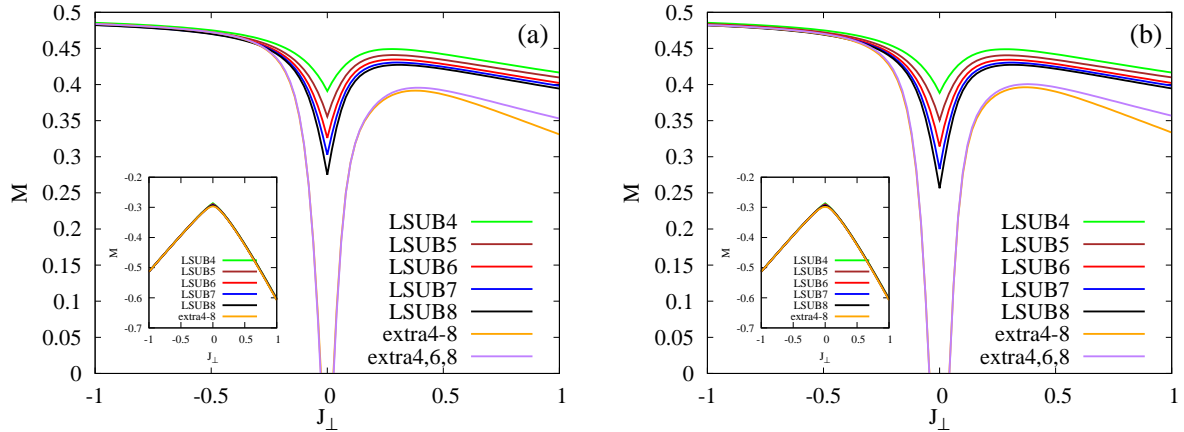


Figure 1. CCM-LSUB m as well as extrapolated GS sublattice magnetization M for $\Delta = 0$ (XY limit) using (a) the $\sqrt{3} \times \sqrt{3}$ reference state and (b) the $q = 0$ reference state as a function of the ILC J_\perp . The labels ‘extra4-8’ and ‘extra4,6,8’ correspond to the extrapolation schemes I and II, respectively (see main text). Inset: Corresponding data for the GS energy e_0 . Note that for e_0 the results of both extrapolation schemes are practically identical, and, therefore we show only extrapolated data for scheme I.

3. Results

We start with a discussion of the most anisotropic XY limit, i.e., $\Delta = 0$. For that case we present in Figs. 1(a) and (b) the full set of LSUB m data as well as the extrapolated data for the order parameter M (main panels) and also for the GS energy per spin $e_0 = E_0/N$ (insets) for the $\sqrt{3} \times \sqrt{3}$ and $q = 0$ reference states, respectively. (Note that corresponding figures for $\Delta = 1$ can be found in [52].) It is evident that e_0 converges quickly as the level m of the LSUB m approximation increases. As a result, the extrapolation with leading order $1/m^2$ is very accurate, as it has been demonstrated in many cases, where data from other precise methods are available to compare with, see, e.g., Refs. [12,30,65]. Obviously, the shape of the curves and the magnitude of the energies are very similar for both states, where from [30] it is known that for the 2D XY model, i.e., at $J_\perp = 0$ and $\Delta = 0$, the $q = 0$ state has slightly lower energy. As found previously [18,29–31,34], M is zero for $J_\perp = 0$. For $\Delta = 0$ the critical ILCs, where magnetic LRO sets in, are : $J_\perp^c = -0.027$, $J_\perp^c = +0.026$ ($\sqrt{3} \times \sqrt{3}$ state) and $J_\perp^c = -0.044$, $J_\perp^c = +0.042$ ($q = 0$ state) when using scheme I, and, $J_\perp^c = -0.028$, $J_\perp^c = +0.028$ ($\sqrt{3} \times \sqrt{3}$ state) and $J_\perp^c = -0.044$, $J_\perp^c = +0.042$ ($q = 0$ state) when using scheme II. Corresponding values for $\Delta = 1$ are [52]: $J_\perp^c = -0.100$, $J_\perp^c = +0.102$ ($\sqrt{3} \times \sqrt{3}$ state) and $J_\perp^c = -0.154$, $J_\perp^c = +0.151$ ($q = 0$ state) for scheme I, and, $J_\perp^c = -0.104$, $J_\perp^c = +0.110$ ($\sqrt{3} \times \sqrt{3}$ state) and $J_\perp^c = -0.135$, $J_\perp^c = +0.130$ ($q = 0$ state) for scheme II. We may conclude that (i) there is a good agreement of the critical ILCs obtained by both extrapolation schemes, (ii) the spin-liquid region survives a finite ILC even in the XY limit, and (iii) due to anisotropy this region becomes noticeably smaller.

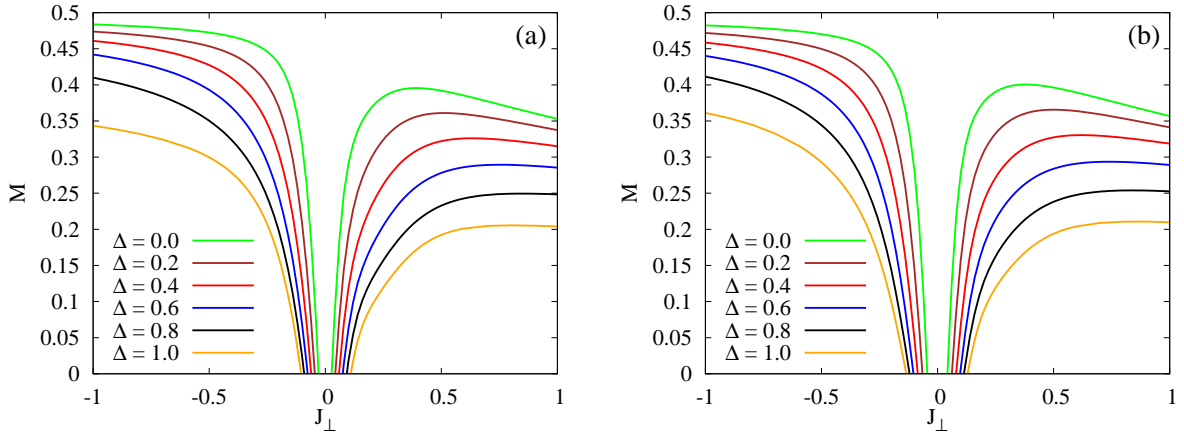


Figure 2. Extrapolated GS sublattice magnetization M (scheme II) for various values of the anisotropy parameter Δ using (a) the $\sqrt{3} \times \sqrt{3}$ reference state and (b) the $q = 0$ reference state as a function of the ILC J_\perp .

In the next figure we compare the extrapolated magnetic order parameter M for several values of the anisotropy parameter Δ , see Figs. 2 (a) and (b). Because (at least for small values of the $|J_\perp|$) both extrapolation schemes lead to very similar results, for convenience, we show only the data of scheme II. The critical values of the ILC, where magnetic LRO sets in, are listed for both extrapolation schemes in Tables 1 and 2. From those tables and from Figs. 2 (a) and (b) we conclude: (i) for all values $0 \leq \Delta \leq 1$ there is a finite region of magnetic disorder, (ii) this region shrinks monotonously with increasing anisotropy (i.e. with decreasing Δ), (iii) near the critical ILC J_\perp^c the downturn of the magnetic order parameter M becomes very steep as increasing the anisotropy, (iv) in the magnetically ordered phase the sublattice magnetization

Table 1. Critical values J_{\perp}^c of the ILC where magnetic LRO emerges for antiferromagnetic J_{\perp} in dependence on the anisotropy parameter Δ . The labels 'extra4-8' and 'extra4,6,8' correspond to the extrapolation schemes I and II, respectively (see main text).

Δ	$q = 0$		$\sqrt{3} \times \sqrt{3}$	
	extra4,6,8	extra4-8	extra4,6,8	extra4-8
0.0	0.042	0.042	0.028	0.026
0.1	0.052	0.052	0.035	0.032
0.2	0.062	0.062	0.043	0.039
0.3	0.071	0.073	0.052	0.046
0.4	0.080	0.083	0.059	0.053
0.5	0.089	0.094	0.068	0.060
0.6	0.097	0.104	0.076	0.068
0.7	0.106	0.115	0.084	0.076
0.8	0.114	0.127	0.093	0.085
0.9	0.122	0.138	0.101	0.094
1.0	0.130	0.151	0.110	0.102

Table 2. Critical values J_{\perp}^c of the ILC where magnetic LRO emerges for ferromagnetic J_{\perp} in dependence on the anisotropy parameter Δ . The labels 'extra4-8' and 'extra4,6,8' correspond to the extrapolation schemes I and II, respectively (see main text).

Δ	$q = 0$		$\sqrt{3} \times \sqrt{3}$	
	extra4,6,8	extra4-8	extra4,6,8	extra4-8
0.0	-0.044	-0.044	-0.029	-0.027
0.1	-0.055	-0.055	-0.037	-0.034
0.2	-0.065	-0.066	-0.045	-0.040
0.3	-0.075	-0.077	-0.053	-0.047
0.4	-0.085	-0.088	-0.061	-0.054
0.5	-0.094	-0.099	-0.069	-0.062
0.6	-0.104	-0.110	-0.076	-0.070
0.7	-0.112	-0.121	-0.083	-0.077
0.8	-0.120	-0.132	-0.091	-0.085
0.9	-0.128	-0.143	-0.098	-0.093
1.0	-0.135	-0.154	-0.104	-0.100

M grows noticeably as increasing the anisotropy, and, (v) while for ferromagnetic $J_{\perp} < 0$ there is a monotonic increase of M with increasing $|J_{\perp}|$, for $J_{\perp} > 0$ we find a non-monotonic behavior of M . Interestingly, for smaller values of Δ the maximum in M appears at values of the ILC significantly before $J_{\perp} = 1$. For $\Delta = 0$ it is at about $J_{\perp} = 0.36$ for both reference states.

Now we address the question which magnetic GS LRO is selected by quantum fluctuations. As we know from previous studies, the mechanism of quantum selection of the GS LRO in the KAFM is very subtle, and the energy difference between competing states can be very small [28, 30, 52]. The CCM approach in high orders of approximation provides an accurate tool

to compare these energies. Thus, the CCM for $s > 1/2$ yields the correct quantum selection of the $\sqrt{3} \times \sqrt{3}$ GS vs. the $q = 0$ GS as obtained by non-linear spin-wave theory [12]. A direct comparison of CCM and non-linear spin-wave data for energy differences (which are also of the order $10^{-3} \dots 10^{-4}$) for the XXZ KAFM for large s , given in Fig. 3 of Ref. [30], demonstrates that both independent approaches agree very well. In Fig. 3 we show our results for the energy difference $\delta e = e_0^{\sqrt{3} \times \sqrt{3}} - e_0^{q=0}$ as a function of J_\perp for various values of Δ . We mention first that both extrapolation schemes I and II yield consistent results for δe . Therefore, for convenience, we show in Fig. 3 only the data of scheme II. From Fig. 3 it is evident that at low values of $|J_\perp|$ the $q = 0$ reference state yields lower energy, i.e. $\delta e > 0$. That is consistent with Refs. [12] and [30], where the case $J_\perp = 0$ was studied. For ferro- as well as antiferromagnetic ILC δe is still positive at those values of J_\perp , where the sublattice magnetizations $M_{\sqrt{3} \times \sqrt{3}}$ or $M_{q=0}$ become larger than zero. Hence, our results demonstrate that a magnetic disorder-to-order transition to $q = 0$ LRO takes place at J_\perp^c as listed in columns 2 and 3 of Tables 1 and 2. We mention that the quantum selection of the $q = 0$ GS LRO is contrary to the semi-classical large- s selection of the $\sqrt{3} \times \sqrt{3}$ LRO found for the 2D spin- s KAFM. As already found for the isotropic case, $\Delta = 1$, further increasing the strength of J_\perp can lead to a second transition at J_\perp^t from $q = 0$ to $\sqrt{3} \times \sqrt{3}$ LRO determined by the change of sign of δe . Here, we find such a second transition for all values Δ on the antiferromagnetic side, $J_\perp > 0$. On the ferromagnetic side a second transition is found only for $\Delta = 1, 0.9, 0.8$. For lower values of Δ the energy difference δe remains positive. However, it is in order to mention that already for $\Delta < 0.9$ our CCM result for δe becomes extremely small as $J_\perp \rightarrow -1$, and the $q = 0$ and $\sqrt{3} \times \sqrt{3}$ states are practically degenerate. We list our results for the position of the second transition between $q = 0$ and $\sqrt{3} \times \sqrt{3}$ magnetic LRO in Table 3. Summarizing our data for the transitions at J_\perp^c and J_\perp^t as given in Tables 1-3 we present in Fig. 4 a sketch of a phase diagram of the stacked $s = 1/2$ XXZ KAFM with easy-plane anisotropy in the layers.

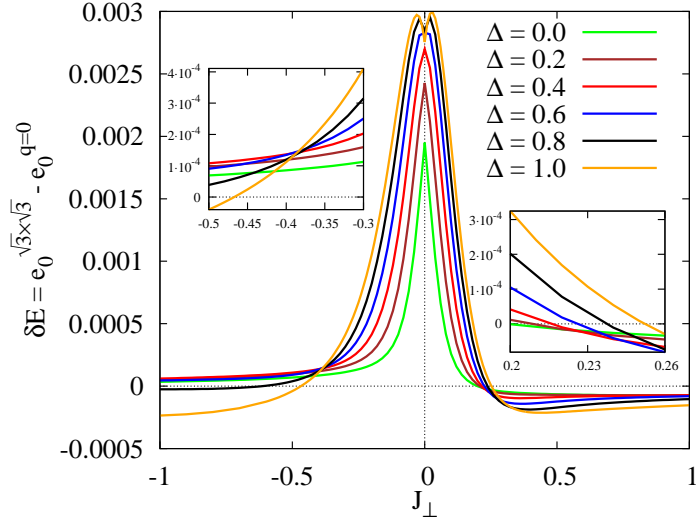


Figure 3. Difference $\delta e = e_0^{\sqrt{3} \times \sqrt{3}} - e_0^{q=0}$ of the extrapolated GS energies (using scheme II) of the $\sqrt{3} \times \sqrt{3}$ and the $q = 0$ states as a function of the ILC J_\perp . In the insets we show δe in an enlarged scale for those regions of J_\perp , where δe changes its sign.

4. Concluding remarks

A main finding of our paper is that the spin-liquid GS present for the strictly 2D $s = 1/2$ XXZ KAFM survives a finite ILC even in the XY limit of maximal easy-plane anisotropy. If the ILC becomes large enough (about 15% of the intralayer coupling in our model system for the isotropic Heisenberg case and about 4.5% for coupled XY kagome layers) magnetic LRO can be

Table 3. Transition points J_{\perp}^t between $q = 0$ and $\sqrt{3} \times \sqrt{3}$ magnetic LRO. For $\Delta < 0.75$ and ferromagnetic J_{\perp} we do find a transition.

Δ	AFM		FM	
	extra4,6,8	extra4-8	extra4,6,8	extra4-8
0.0	0.199	0.220	—	—
0.1	0.205	0.234	—	—
0.2	0.209	0.242	—	—
0.3	0.212	0.248	—	—
0.4	0.216	0.254	—	—
0.5	0.220	0.259	—	—
0.6	0.226	0.265	—	—
0.7	0.232	0.273	—	—
0.8	0.238	0.282	-0.614	-0.562
0.9	0.245	0.294	-0.502	-0.466
1.0	0.252	0.310	-0.467	-0.435

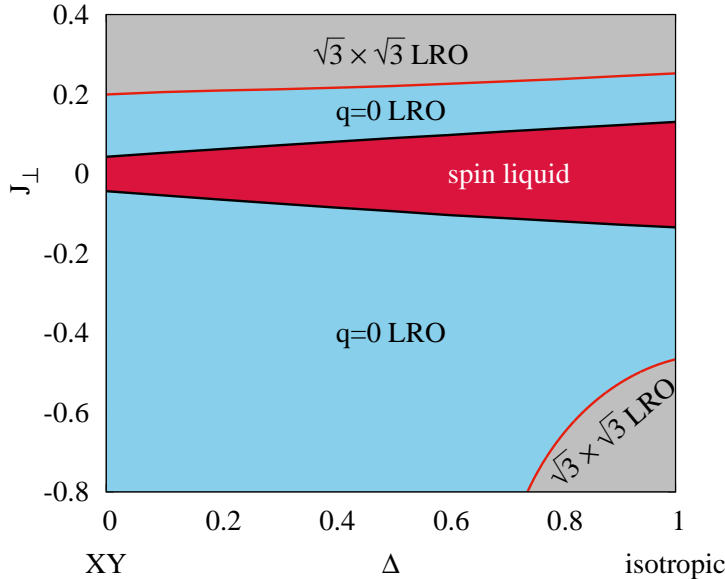


Figure 4. Sketch of a phase diagram of the stacked $s = 1/2$ kagome antiferromagnet with easy-plane anisotropy in the layers.

established, where the $q = 0$ symmetry is favorable, if J_{\perp} is of moderate strength. The quantum selection of the $\sqrt{3} \times \sqrt{3}$ magnetic LRO versus the $q = 0$ magnetic LRO found for larger values of antiferromagnetic J_{\perp} and also for ferromagnetic J_{\perp} near the isotropic limit is related to a very small energy gain. Thus, in real compounds even very small additional terms in the relevant spin Hamiltonian such as further distance exchange couplings may therefore be more relevant.

Let us finally discuss the relevance of our results for experiments on kagome compounds. As it has been pointed out very recently, through isoelectronic substitution in barlowite this kagome compound is related to our model system of stacked (unshifted) kagome layers [51]. The finding that the spin-liquid phase survives a finite ILC is relevant for the modified barlowite system, where an ILC of about 6-7% of the intralayer coupling was predicted [51]. The relation to herbertsmithite, for which an ILC of about 5% was determined [76], is only on a qualitative

level, since here the kagome layers are shifted from layer to layer. The observation of $q = 0$ magnetic order in $\text{Cs}_2\text{Cu}_3\text{SnF}_{12}$ reported in [77, 78] is also in qualitative agreement with our findings for the GS selection. We also mention that anisotropic kagome antiferromagnets have relevance for the experimental research, see, e.g., Refs. [77, 79–81]. Moreover, anisotropic spin models are of great interest with respect to engineering models of quantum magnetism on optical lattices, see, e.g., [82–84].

References

- [1] R. R. P. Singh and D. A. Huse, *Phys. Rev. Lett.* **68**, 1766 (1992).
- [2] C. Waldtmann et al., *Eur. Phys. J. B* **2**, 501 (1998).
- [3] S. Capponi, A. Läuchli, and M. Mambrini, *Phys. Rev. B* **70**, 104424 (2004).
- [4] R. R. P. Singh and D. A. Huse, *Phys. Rev. B* **76**, 180407(R) (2007).
- [5] P. Sindzingre and C. Lhuillier, *Europhys. Lett.* **88**, 27009 (2009).
- [6] G. Evenbly and G. Vidal, *Phys. Rev. Lett.* **104**, 187203 (2010).
- [7] S. Yan, D. A. Huse, and S. R. White, *Science* **332**, 1173 (2011).
- [8] H. Nakano and T. Sakai, *J. Phys. Soc. Jpn.* **80**, 053704 (2011).
- [9] A. M. Läuchli, J. Sudan, and E. S. Sørensen, *Phys. Rev. B* **83**, 212401 (2011).
- [10] Y.-M. Lu, Y. Ran, and P. A. Lee, *Phys. Rev. B* **83**, 224413 (2011).
- [11] Y. Iqbal, F. Becca, and D. Poilblanc, *Phys. Rev. B* **84**, 020407(R) (2011).
- [12] O. Götze et al., *Phys. Rev. B* **84**, 224428 (2011).
- [13] S. Depenbrock, I. P. McCulloch, and U. Schollwöck, *Phys. Rev. Lett.* **109**, 067201 (2012).
- [14] Y. Iqbal, F. Becca, S. Sorella, D. Poilblanc, *Phys. Rev. B* **87**, 060405(R) (2013).
- [15] I. Rousochatzakis, R. Moessner, J. van den Brink, *Phys. Rev. B* **88**, 195109 (2013).
- [16] Z. Y. Xie et al., *Phys. Rev. X* **4**, 011025 (2014).
- [17] I. Rousochatzakis, Y. Wan, O. Tchernyshyov, and F. Mila, *Phys. Rev. B* **90**, 100406(R) (2014).
- [18] W.-J. Hu, S.S. Gong, F. Becca, and D.N. Sheng, *Phys. Rev. B* **92**, 2015(R) (2015).
- [19] M. P. Zaletel and A. Vishwanath, *Phys. Rev. Lett.* **114**, 077201 (2015).
- [20] J. Oitmaa and R. R. P. Singh, *Phys. Rev. B* **93**, 014424 (2016).
- [21] T. Liu, W. Li, A. Weichselbaum, J. von Delft, and G. Su, *Phys. Rev. B* **91**, 060403 (2015).
- [22] H. J. Changlani and A. M. Läuchli, *Phys. Rev. B* **91**, 100407 (2015).
- [23] S. Nishimoto and M. Nakamura, *Phys. Rev. B* **92**, 140412(R) (2015).
- [24] W. Li, A. Weichselbaum, J. von Delft, and H.-H. Tu, *Phys. Rev. B* **91**, 224414 (2015).
- [25] Tao Liu, Wei Li, Gang Su, arXiv:1603.01935v1.
- [26] O. Cepas, C. M. Fong, P. W. Leung, C. Lhuillier, *Phys. Rev. B* **78**, 140405(R) (2008).
- [27] I. Rousochatzakis, S. R. Manmana, A. M. Läuchli, B. Normand, and F. Mila, *Phys. Rev. B* **79**, 214415 (2009).
- [28] A. L. Chernyshev and M. E. Zhitomirsky, *Phys. Rev. Lett.* **113**, 237202 (2014).
- [29] Y.C. He and Y. Chen, *Phys. Rev. Lett.* **114**, 037201 (2015).
- [30] O. Götze and J. Richter, *Phys. Rev. B* **91**, 104402 (2015).
- [31] W. Zhu, S. S. Gong, and D. N. Sheng, *Phys. Rev. B* **92**, 014424 (2015).
- [32] K. Kumar, K. Sun, and E. Fradkin, *Phys. Rev. B* **92**, 094433 (2015).
- [33] A. L. Chernyshev and M. E. Zhitomirsky, *Phys. Rev. B* **92**, 144415 (2015).
- [34] K. Essafi, O. Benton, and L.D.C. Jaubert, *Nat. Commun.* **7**, 10297 (2016).
- [35] J.-C. Domenge, P. Sindzingre, C. Lhuillier, and L. Pierre, *Phys. Rev. B* **72**, 024433 (2005).
- [36] O. Janson, J. Richter, and H. Rosner, *Phys. Rev. Lett.* **101**, 106403 (2008).
- [37] R.F. Bishop, P.H.Y. Li, D.J.J. Farnell, and C.E. Campbell, *Phys. Rev. B* **82**, 104406 (2010).
- [38] T. Tay and O. I. Motrunich, *Phys. Rev. B* **84**, 020404(R) (2011).
- [39] P.H.Y. Li, R.F. Bishop, C.E. Campbell, D.J.J. Farnell, O. Götze, and J. Richter, *Phys. Rev. B* **86**, 214403 (2012).
- [40] H.-C. Jiang, Z. Wang, and L. Balents, *Nature Physics* **8**, 902 (2012).
- [41] B. Bauer et al., *Nature Communications* **5**, 5137 (2014).
- [42] R. Suttner, C. Platt, J. Reuther, and R. Thomale, *Phys. Rev. B* **89**, 020408(R) (2014).
- [43] W.J. Hu, W. Zhu, Y. Zhang, S.S. Gong, F. Becca, and D.N. Sheng, *Phys. Rev. B* **91**, 041124(R) (2015).
- [44] S.S. Gong, W. Zhu, L. Balents, and D. N. Sheng, *Phys. Rev. B* **91**, 075112 (2015).
- [45] F. Kolley, S. Depenbrock, I. P. McCulloch, U. Schollwöck, and V. Alba, *Phys. Rev. B* **91**, 104418 (2015).
- [46] S. Bieri, L. Messio, B. Bernu, and C. Lhuillier, *Phys. Rev. B* **92**, 060407(R) (2015).
- [47] A. Wietek, A. Sterdyniak, and A. M. Läuchli, *Phys. Rev. B* **92**, 125122 (2015).
- [48] Y. Iqbal et al., *Phys. Rev. B* **92**, 220404 (2015).

- [49] T.-H. Han, J. Singleton, J. A. Schlueter, *Phys. Rev. Lett.* **113**, 227203 (2014).
- [50] H. O. Jeschke et al., *Phys. Rev. B* **92**, 094417 (2015).
- [51] D. Guterding, R. Valenti, and H. O. Jeschke, arXiv:1605.08162.
- [52] O. Götze and J. Richter, *Europhys. Lett. (EPL)* **114**, 67004 (2016).
- [53] D. Schmalfuß, R. Darradi, J. Richter, J. Schulenburg, and D. Ihle, *Phys. Rev. Lett.* **97**, 157201 (2006).
- [54] R. Darradi, O. Derzhko, R. Zinke, J. Schulenburg, S.E. Krüger and J. Richter, *Phys. Rev. B* **78**, 214415 (2008).
- [55] R. Zinke, J. Schulenburg, and J. Richter, *Eur. Phys. J. B* **61**, 147 (2008).
- [56] D.J.J. Farnell, R. Zinke, J. Schulenburg, and J. Richter, *J. Phys.: Cond. Matter* **21**, 406002 (2009).
- [57] J. Richter, R. Darradi, J. Schulenburg, D.J.J. Farnell, and H. Rosner, *Phys. Rev. B* **81**, 174429 (2010).
- [58] D.J.J. Farnell et al., *Phys. Rev. B* **84**, 012403 (2011).
- [59] D.J.J. Farnell et al., *Phys. Rev. B* **89**, 184407 (2014).
- [60] R. F. Bishop, P. H. Y. Li, and C. E. Campbell, *Phys. Rev. B* **89**, 214413 (2014).
- [61] J.-J. Jiang, Y.-J. Liu, F. Tang, C.-H. Yang, and Y.-B. Sheng, *Physica B: Cond. Mat.* **463**, 30 (2015).
- [62] J. Richter, R. Zinke, D.J.J. Farnell, *Eur. Phys. J. B* **88**, 2 (2015).
- [63] P. H. Y. Li, R. F. Bishop, and C. E. Campbell, *Phys. Rev. B* **91**, 014426 (2015).
- [64] R.F. Bishop, in *Microscopic Quantum Many-Body Theories and Their Applications*, edited by J. Navarro and A. Polls, *Lecture Notes in Physics* **510** (Springer, Berlin, 1998), p.1.
- [65] D.J.J. Farnell and R.F. Bishop, in *Quantum Magnetism, Lecture Notes in Physics* **645**, 307 (2004)
- [66] R.F. Bishop, D.J.J. Farnell, S.E. Krüger, J.B. Parkinson, *J. Phys.: Condens. Matter* **12**, 6887 (2000).
- [67] M. Roger and J.H. Hetherington, *Phys. Rev. B* **41**, 200 (1990).
- [68] R.F. Bishop, J.B. Parkinson, and Y. Xian, *Phys. Rev. B* **43**, R13782 (1991); *Phys. Rev. B* **44**, 9425 (1991).
- [69] C. Zeng, D.J.J. Farnell, and R.F. Bishop, *J. Stat. Phys.* **90**, 327 (1998).
- [70] A. B. Harris, C. Kallin and A. J. Berlinsky, *Phys. Rev. B* **45**, 2899 (1992).
- [71] S. Sachdev, *Phys. Rev. B* **45**, 12377 (1992).
- [72] A. Chubukov, *Phys. Rev. Lett.* **69**, 832 (1992).
- [73] C. L. Henley and E. P. Chan, *J. Magn. Magn. Mater.* **140-144**, 1693 (1995).
- [74] For the numerical calculation we use the program package ‘The crystallographic CCM’ (D.J.J. Farnell and J. Schulenburg).
- [75] D. J. J. Farnell and R. F. Bishop, *Int. J. Mod. Phys. B* **22**, 3369 (2008).
- [76] O. Janson, *DFT based microscopic magnetic modeling for low-dimensional spin systems*, Ph.D. thesis, Technische Universität Dresden (2012)
- [77] T. Ono, K. Matan, Y. Nambu, T. J. Sato, K. Katayama, S. Hirata, and H. Tanaka, *J. Phys. Soc. Jpn.* **83**, 043701 (2014); K. Katayama, N. Kurita, H. Tanaka, arXiv:1412.5770.
- [78] K. Katayama, N. Kurita, and H. Tanaka, *Phys. Rev. B* **91**, 214429 (2015).
- [79] K. Matan, D. Grohol, D. G. Nocera, T. Yildirim, A. B. Harris, S. H. Lee, S. E. Nagler, and Y. S. Lee, *Phys. Rev. Lett.* **96**, 247201 (2006).
- [80] A. Zorko, S. Nellutla, J. van Tol, L. C. Brunel, F. Bert, F. Duc, J.-C. Trombe, M. A. de Vries, A. Harrison, and P. Mendels, *Phys. Rev. Lett.* **101**, 026405 (2008).
- [81] A. Zorko, F. Bert, A. Ozarowski, J. van Tol, D. Boldrin, A. S. Wills, and P. Mendels, *Phys. Rev. B* **88**, 144419 (2013).
- [82] S. V. Isakov, S. Wessel, R. G. Melko, K. Sengupta, Yong Baek Kim, *Phys. Rev. Lett.* **97**, 147202 (2006).
- [83] I. Bloch, J. Dalibard, and W. Zwerger, *Rev. Mod. Phys.* **80**, 885 (2008).
- [84] J. Struck, M. Weinberg, C. Ölschläger, P. Windpassinger, J. Simonet, K. Sengstock, R. Höppner, P. Hauke, A. Eckardt, M. Lewenstein, and L. Mathey, *Nature Physics* **9**, 738 (2013).